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High potential of calcareous tufas for integrative multidisciplinary studies and prospects for archaeology in Europe

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ABSTRACT

Calcareous tufas are continental carbonates from open-air conditions, specific to wet and warm periods. They contain abundant remains of fauna and flora fossilised *in situ* and may accumulate regularly over thousands of years offering high stratigraphic resolution for palaeoenvironmental reconstructions. As they are mainly comprised of calcite, tufas allow direct and precise dating as well as geochemical reconstructions of past climates. Additionally, recent investigations have highlighted their strong potential for archaeology as several studied sequences provided high quality record of *in situ* prehistoric levels. Tufas are thus a unique archive in continental areas for development of synergic multidisciplinary investigations of past human societies and associated environmental and climate evolution. We emphasise that calcareous tufa are key-deposits to investigate human–environment–climate interactions during interglacial periods, from Lower Palaeolithic to Antiquity, in Europe.

1. Introduction

Calcareous tufas are continental carbonate rocks deposited in open-air conditions in streams and rivers, widely observed in fluvial areas with calcareous bedrock (Pentecost, 1995). Tufas result from dissolution of the carbonate content of the bedrock in the aquifer, favoured by soil and forest development and the resulting groundwater acidification. When water then flows from the spring, variations of its physicochemical parameters and activity of the instream vegetation (especially cyanobacteria, mosses and algae) lead to rapid calcite precipitation. Detrital and calcite accumulation over years (mm to cm/yr; Pentecost, 2005; Gradziński, 2010; Vázquez-Urbez et al., 2010) generally results to fine, porous, and slightly layered to lenticular deposits (Casanova, 1981; Pedley, 1990; Ford and Pedley, 1996). Those characteristics induce (1) that tufas are specific to wet and warm periods (Pentecost, 2005), which allow significant groundwater flows, deep soil forest development and optimal conditions (warm and sunny environments) for cyanobacteria and algae growing, (2) that they contain abundant remains of fauna and flora fossilised *in situ* (Capezzuoli et al., 2014), and (3) that they may accumulate regularly over thousands of years offering a high stratigraphic resolution for environmental and climatic reconstructions (Pentecost, 2005).

Large fluvial basins running on calcareous geological formations are common in Europe (Thames Basin, Paris and Aquitaine Basins, Iberian range, etc.), which allows a wide geographical distribution of tufa formations (Fig. 1). These fluvial systems are often characterised by terrace sequences. The Seine, Somme, Yonne (France), Thames (UK), Rhine or Meuse (Germany) valleys present at least ten stepped terraces covering the last million years (Antoine, 1994; Lautridou et al., 1999; Chaussé et al., 2004; Bridgland et al., 2004). Multidisciplinary studies, combining stratigraphy, sedimentology, palaeontology and absolute dating, have demonstrated that Quaternary climate cyclical variations that modified fluvial environments were the major factor leading to stepped systems. Fluvial terraces thus result from the glacial/interglacial morphosedimentary variations; their succession correlates to Marine Isotopic Stage (MIS) cyclicity.

In these terrace systems, as for example in the Somme valley (France), temperate periods were initially observed in silts at the top of the fluvial sequences. In these deposits, bioindicators record early or late interglacial environmental conditions whereas climatic optima were not observed (Antoine et al., 2000, 2003). In the early 2000's, a new malacological study of the historical sequence of Saint-Acheul (Somme Valley, France; Fig. 1) demonstrated full

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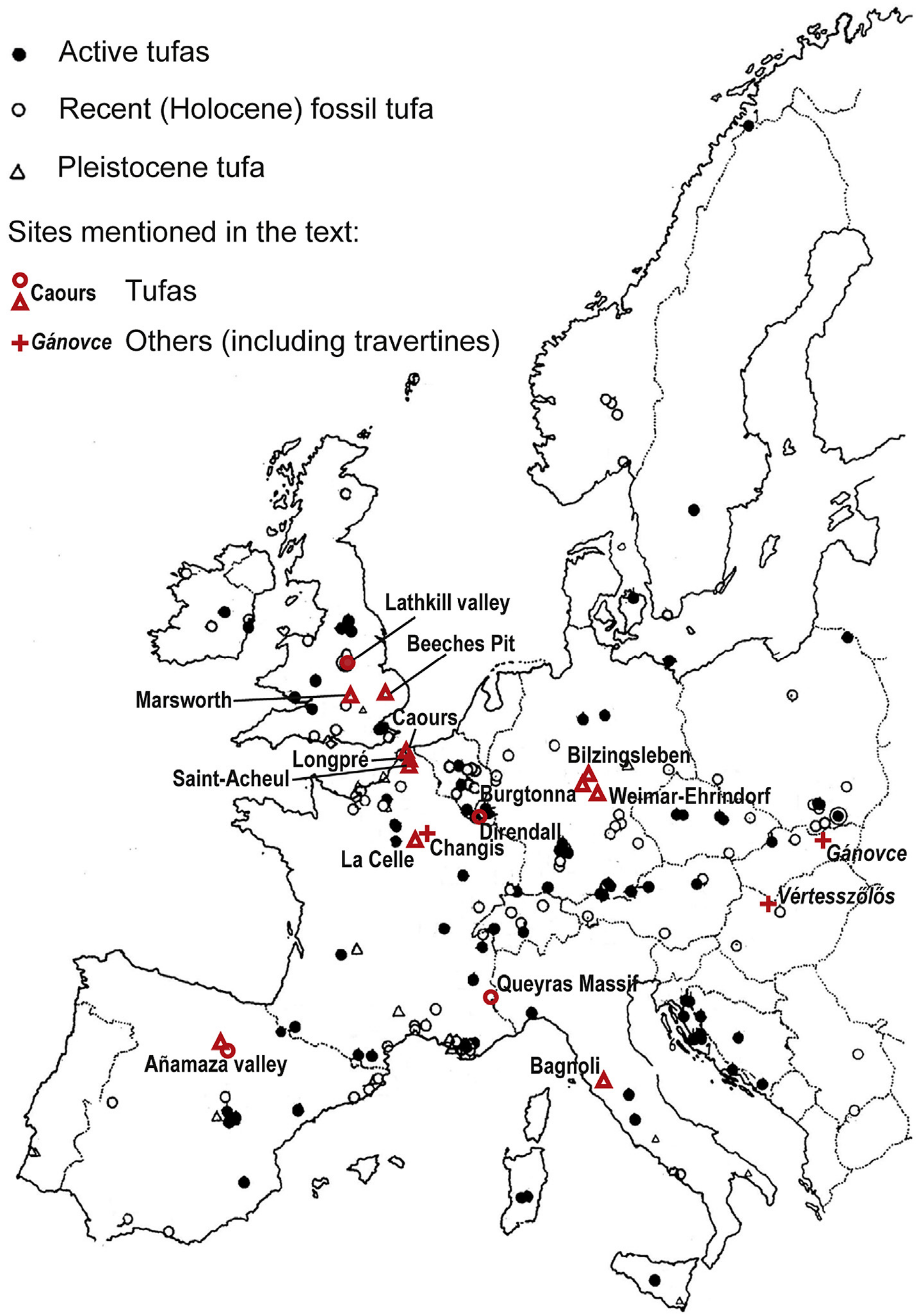

Fig. 1. Distribution of calcareous tufas in Europe (modified after Pentecost, 1995) and localisation of tufa sites mentioned in text. For detailed information on each site, refer to the following publications: **Añamaza valley**, Holocene to MIS 5, Arenas et al., 2014; **Bagnoli**, MIS 5, Bertini et al., 2014; **Beeches Pit**, MIS 11, Preece et al., 2006, 2007; **Caours**, MIS 5e, Antoine et al., 2006, Dabkowski, 2011, Dabkowski et al., 2010, 2011, submitted for publication, Locht et al., 2009; **Direndall**, Holocene, Meyrick, 2002; **Gánovce**, MIS 5, Vlček, 1955, 1995; **La Celle**, MIS 11, Dabkowski, 2011, Dabkowski et al., 2011, 2012; Limondin-Lozouet et al., 2006, 2010, **Lathkill valley**, Holocene and modern, Andrews et al., 1994, Pedley et al., 2000; **Marsworth**, MIS 11, Green et al., 1984; **Queyras Massif**, Holocene, Ali et al., 2004; **Saint-Acheul**, MIS 11, Antoine and Limondin-Lozouet, 2004, Limondin-Lozouet and Antoine, 2006; **Thuringia** (Bilzingsleben, Burgtonna, Weimar-Ehringsdorf), MIS 5 and MIS 11, Kahlke, 2002, Kahlke and Wunderlich, 2003, Meyrick and Schreve, 2002, Müller and Pasda, 2011; **Vértesszöllös**, MIS 11, Pentecost, 2005.

temperate conditions in a tufa unit characterised by forest thermophilous fauna and assigned to the MIS 11 climatic optimum (Antoine and Limondin-Lozouet, 2004; Limondin-Lozouet and Antoine, 2006). Calcareous tufas actually appear to be the only deposits recording the interglacial optima in fluvial contexts (Antoine et al., 2003). Regained interest in tufas over the last decade involved different research fields such as sedimentology, palaeontology, hydrology, and geochemistry in order to reconstruct past environmental succession and climate of Pleistocene temperate periods.

Recent investigations have highlighted the strong potential of calcareous tufa for archaeology as several studied sequences also provided high quality record of prehistoric settlements. We present here some tufa specificities that might have an extensive potential for archaeology, illustrated by selected examples from fossil tufa sequences generally associated to human occupations.

2. Tufa deposits and resulting *in situ* preservation of archaeological informations

The morphology, granulometry and composition of tufa facies mainly depends on the geomorphological position of the deposit, the presence/absence of vegetation and algae and the speed of the water flow (Casanova, 1981; Pedley, 1990; Pentecost, 2005; Capezzuoli et al., 2014). Fluvial, paludal and lacustrine tufas might be separated even if each natural system is generally composed of more than one model system (Ford and Pedley, 1996). As fluvial areas have been favoured for human settlements in Europe since the Early Palaeolithic (Antoine et al., 2003, 2010; Ashton et al., 2006; Bridgland et al., 2006), tufa associated to fluvial contexts are the most of interest here.

The fluvial model (Pedley, 1990) includes braided fluvial, barrage and perched springline systems, according to the position of the tufa deposits along the river profile (Ford and Pedley, 1996). Shallow braided river tufas are dominated by oncolithes and phytoclastic lenses mixed with various detrital materials (Casanova, 1981; Ford and Pedley, 1996). Traverse barriers across stream result of barrage systems dominated by hard tufa facies that builds up laminar to low domes and encrusts mosses, algae and various in place plant. Pools upstream of the barrages are generally shallow and dominated by lime mud and organic deposits (Pedley et al., 1996). These stream bed deposits are very unlikely to present *in situ* archaeological sites but can preserved natural accumulations of various palaeontological remains. The perched springline models (or slope systems) are fed by springs or stream system resurgences at a higher point on a pre-existing valley side. They are characterised by lobate or multilobate flat to slightly convex deposits thickening away from sources (Ford and Pedley, 1996). Perched springline tufas are never associated with significant bodies of standing water and are generally composed of friable, organic-rich, low dynamic deposits with commonly preserved palaeosoils (Pedley et al., 2003). Slopes near springs or river banks are also areas where human populations are the most likely to settle. The perched springline tufa systems are therefore very likely to preserve *in situ* archaeological artefacts and palaeontological remains.

Natural fluvial tufa systems are generally composed of combined braided fluvial, barrage and slope tufa deposits depending on the morphology of the valley and evolution of the local hydrodynamics (mainly the water table). Fluvial tufas are also likely to interconnect with other fluvial deposits as clays, silts or sands, and peat deposits. Such composite systems have been described in details in some recent to Pleistocene sites: e.g. the Lathkill valley (Derbyshire, UK; Pedley et al., 2000), the Añamaza valley (Iberian Range, Spain; Arenas et al., 2014) or Caours (Somme valley, France; Antoine et al., 2006; Locht et al., 2009, Fig. 1).

Fluvial areas and especially river banks and slopes near springs were preferentially occupied by humans since the Early Palaeolithic (Antoine et al., 2003, 2010; Ashton et al., 2006; Bridgland et al., 2006). These environments and associated tufa deposits, mainly perched spingline (slope) systems, are thus key-areas for archaeological investigations. Human settlements would be naturally favoured on waterless surface associated to low sedimentation rate and soil development. In tufa contexts, archaeological levels are commonly preserved in units characterised by fine greyish deposits indicating low deposition rate and early soil development. These units generally comprise malacofaunes indicating stabilisation of the surrounding environment (forest maximal development). Such palaeosoils are common in slope tufa deposits (Pedley et al., 2003). For example, they have been clearly observed in the Caours tufa (Somme Basin, France; Fig. 1) where the two upper Middle Palaeolithic levels are preserved in greyish fine tufa units which are associated to terrestrial malacofaunes and show microfacies in thin section characterising dry phases (Antoine et al., 2006; Dabkowski et al., 2010).

After abandonment, human settlements can be uncovered by tufa deposits especially when the water dynamics has changed allowing faster sedimentation. High calcite accumulation rate would rapidly seal archaeological levels in tufa or associated sediments (such as fluvial clay or peat deposits) and are therefore very likely to be preserved *in situ*. The conservation of 400 ka years old hearth structures in Beeches Pit (Figs. 1 and 2) is a remarkable example of the quality of archaeological surfaces that can be uncovered if carefully excavated. Burned structures associated to lithic artefacts are here preserved at the surface of silty deposits rapidly sealed by tufa deposition (Preece et al., 2006).

In such well preserved contexts, different activity areas (fire, knapping, butchery, etc.) can be mapped precisely, as left by the early humans, which allows their spatial organisation and possible interaction within the site being studied. For example, the Caours tufa deposits (Somme Basin, Northern France; MIS 5e; Fig. 1) have allowed an excellent preservation of at least four Middle Palaeolithic levels (Antoine et al., 2006). One of them, (level N3) has been preserved in a peat layer rapidly sealed by calcareous tufa. It comprises 1113 faunal remains (red deer, fallow deer, roe deer, aurochs, etc.) and 319 lithic artefacts (association of unipolar, Levallois and discoidal production systems). Mapping clearly shows that the formers are more abundant in the north-western part of the excavation (Fig. 3). Regarding the spatial concentration of bone artefacts and their features (cut marks and breakage patterns), this north-eastern area is interpreted as a butchery area. In the central part, a little knapping place was discovered *in situ* (Fig. 3). Lithic tools appear to be produced here for direct use on the animal carcasses. Finally, the south-eastern part of the excavation has few or none artefacts. It was apparently not occupied by humans and must correspond to a humid depression zone (Locht et al., 2009).

3. Environmental data recorded in tufas

3.1. Large mammal (including human) and vertebrate remains

In the case of natural accumulation, large mammals are commonly preserved in anatomical connection and most of the skeleton is present as seen in Thuringia basin sites in Central Germany (Burgtonna, Weimar-Ehringdorf, Bilzingsleben; Kahlke, 2002; Kahlke and Wunderlich, 2003; Müller and Pasda, 2011,

Fig. 2. Photograph of fire hearths including burned flints at Beeches Pit (Area AH), showing an area of 3 m by 2 m viewed from the west (modified after Preece et al., 2006).

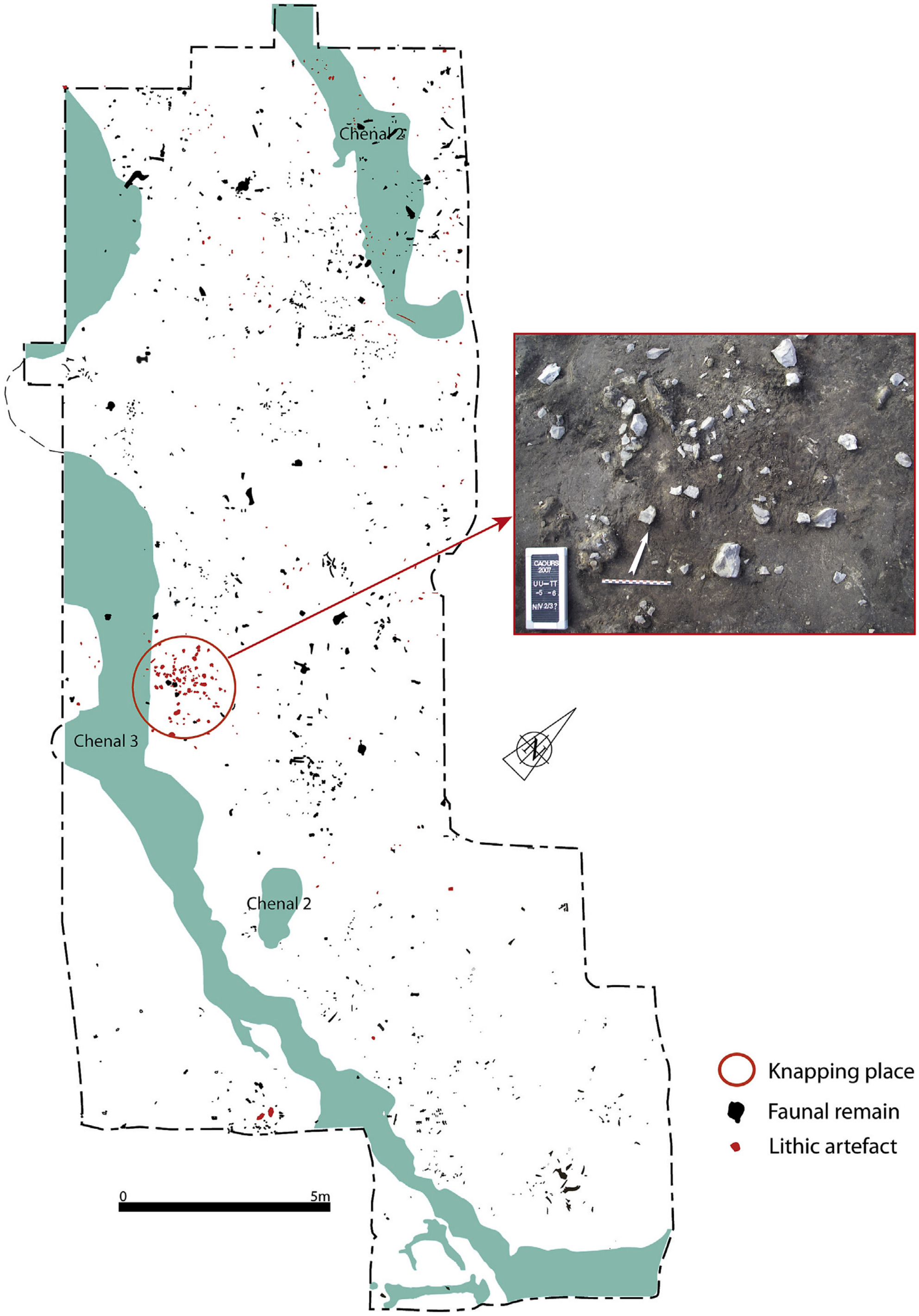

Fig. 3. Mapping of archaeological artefacts form level N3 at Caours (Locht et al., 2009) and detail of the knapping place preserved *in situ* in the central area of the excavation (photo: JL. Locht).

Fig. 1). In this area, remains of *Homo heidelbergensis* and *Homo neanderthalensis* have also been found in the tufa deposits, associated to the large mammal fauna (Mania et al., 1994). Human fossils preserved in European tufas remain exceptional. This palaeoanthropological record can be completed by findings in similar open air carbonate deposits, but associated with deep thermal circulating water (hot spring deposits), that should be classified as “travertines” *s. str.* (Capezzuoli et al., 2014). In Gánovce (Slovakia), a Neanderthal skull endocast with some cranial bones still attached was found in a travertine layer dated around 100 ky, which also revealed Middle Paleolithic lithic artefacts (Vlček, 1955, 1995). A cranium fragment of *Homo erectus* was also discovered with stone tools and other mammalian bones in the Vértesszöllös travertine (Hungary), which is probably older than 350 ka (Pentecost, 2005).

In continental carbonate deposits such as travertines and tufas, large non-human mammal remains are yet much more common. Together with a wide range of other vertebrate fossils including small mammals, amphibians, reptiles, birds and fishes that can also

be well preserved in calcareous tufas, they support environmental and paleontological investigations (Pentecost, 2005).

Large mammals typically found in European tufas are, depending on the geological age of the sequence, *Cervus elaphus* (red deer), *Dama* clactoniana and *Dama dama* (fallow deer), *Capreolus capreolus* (roe deer), *Bos primigenius* (aurochs), *Stephanorhinus hemitoechus* (meadow rhinoceros), *Stephanorhinus kirchbergensis* (forest rhinoceros), *Sus scrofa* (wild boar), *Ursus arctos* (brown bear), *Ursus deningeri* (Deninger's bear), *Palaeoloxodon antiquus* (forest elephant), *Macaca sylvanus* (macaque monkey), *Felis sylvestris* (wild cat), *Equus taubachensis* (Taubach horse), *Hippopotamus antiquus* (hippopotamus). These taxa are characteristic of temperate conditions with landscapes dominated by more or less open forest. In Central and Eastern Europe, *Bubalus murrensis* (water buffalo), *Alces latifrons* (broad-antler elk) and *Megaloceros giganteus* (megaloceros) are present while *Bison priscus* (steppe bison) appears more common than aurochs (Schreve and Bridgland, 2002; Auguste, 2009). For each interglacial, a biogeographical gradient is recorded in European tufa deposits from more oceanic conditions in the Western Europe to continental climate in the Central and Eastern part. Additionally, tufa contexts can preserve these large mammal remains in excellent state, which allows, in archaeological context, conservation of human processing features, like cut marks or breaking patterns of long bones.

3.2. *Organic fossil imprints*

High quality data are frequently preserved in tufa sediments as prints of organic remains which have decayed. The exceptional preservation of a Neanderthal skull in the Gánovce travertine (Slovakia) was already mentioned above. This cranium endocast has kept the remaining cranial bones together and preserved the geometry of the skull where bones are missing (Vlček, 1955). Conservation by recrystallisation of the bone structure is very precise as collagen fibrils, bone canaliculi and fibres in muscle insertions have been identified (Vlček, 1995).

Leaf prints appear to be considerably more common in Europe (Pentecost, 2005). The best known Quaternary tufa flora comes from important sites in Central Germany: Weimar-Ehringsdorf, Bilzingsleben and Burgtonna tufas (Thuringia, Germany; Fig. 1) have provided a rich and exceptionally well preserved collection of leaf associated to insect prints (Fig. 4; Meyrick and Schreve, 2002). Some other major European vegetal print collections come from Marsworth (UK; Green et al., 1984; Field, 1993), the Queyras Massif (French Alps; Ali et al., 2004) and La Celle (France; Limondin-Lozouet et al., 2006, Figs. 1 and 4). Processes leading to leaf impressions in tufa have been little studied but observation of active tufa deposits shows that they require fast burial of the vegetal (or any organic) remains by very fine tufa allowing anaerobic conditions and preservation of the organic matter for at least a few years (Pentecost, 2005). Encrustation of living plants is also common and leads to erected (in life position) moulds of mosses, algae, stems and reeds, or even trunks.

After decay of the organic support the remaining calcite mould may provide external details allowing specific identification and thus environmental investigations. As an example, the leaf prints from the MIS 11 La Celle tufa in Northern France (Figs. 1 and 4) provided important information to reconstruct part of the flora contemporaneous of the Early Palaeolithic occupation (Limondin-Lozouet et al., 2006, 2010). The leaf print collection gathered in the late 19th century (Munier-Chalmas Collection housed at the University Paris VI, Jussieu) were recently re-examined and provided one of the best records of the vegetation in NW Europe during the MIS 11 optimum (Jolly-Saad et al., 2006). Several Mediterranean taxa were identified from leaf and fruit prints (Fig. 4), such as fig tree (*Ficus carica*), boxwood (*Buxus sempervirens*) and European nettle tree (*Celtis australis*), which confirms the warm interglacial conditions indicated by the malacological data (Limondin-Lozouet et al., 2010).

3.3. *Long term environmental reconstruction: malacology, ostracology and palynology*

When developed to several tens of centimetres high or more (ideally several metres), calcareous tufa sequences not only provide information contemporaneous with human occupations but also allow reconstruction of the detailed environmental and climatic succession recorded within the deposit.

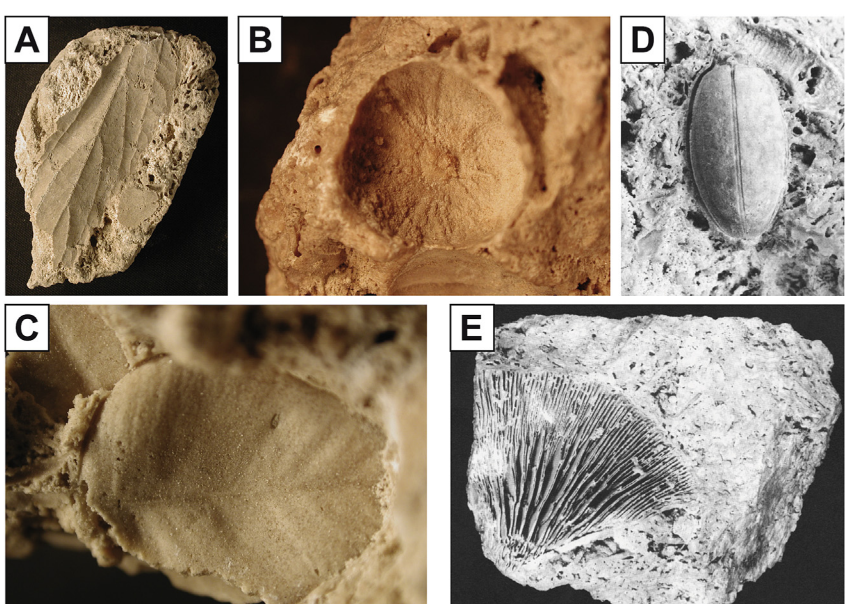

Fig. 4. Organic fossil imprints in tufa. A–C : leaves and fruit from La Celle (A: *Celtis* sp., B: *Fucus* sp., *Tilia* sp.; after Jolly-Saad et al., 2006); D: Wing case from a ground beetle (*Carabus* sp.) from Burgtonna and E: mushroom gills (indeterminate) from Ehrindorf-Weimar (photos T. Korn *in* Meyrick and Schreve, 2002).

Molluscs, and to a less extent ostracod shells, are well represented both in quantity and quality in tufas (especially in soft and fine facies; Fig. 5), which provide favourable conditions for the development and preservation of those communities of invertebrates (Preece, 1991; Carbonel et al., 1988). Molluscs can colonise both terrestrial and aquatic habitats and are strongly dependent on vegetation and so, reflect local environmental conditions. Quaternary malacological successions are therefore suitable for reconstructing environmental variations and corresponding climatic contexts. They have been increasingly investigated for several decades, especially within tufa deposits (e.g. Kerney, 1976; Mania, 1978; Rousseau et al., 1992; Limondin-Lozouet et al., 2006; Preece et al., 2007). Malacofaunes have also proved to be reliable chronological indicators when biogeographical approaches were developed (e.g. Preece, 1991; Keen, 2001; Limondin-Lozouet and Antoine, 2006).

In tufa deposits, the malacological succession from La Celle is an interesting example, which has provided a detailed story of the palaeoenvironmental changes during the interglacial period allocated to MIS 11. Evolution in the composition of mollusc faunas reflects the appearance of forest cover and its progressive extension up to a maximum phase corresponding to the interglacial optima. This last is followed by a terminal environmental episode during which the deciduous forest is replaced by conifer stand (Limondin-Lozouet, 2011). Moreover, the *Lyrodiscus* assemblage recorded in the second half of the tufa formation strengthens the attribution of the tufa deposit to MIS 11, demonstrating the occurrence of a peculiar biotope of humid and temperate forest (Limondin-Lozouet et al., 2006, 2010).

Pollen grains were considered to be little preserved in tufa except for occasional occurrences in fine grained organic levels within Holocene formations (i.e. Fritz, 1976; Geurts, 1976; Preece and Day, 1994; Gauthier *in* Limondin-Lozouet et al., 2005). Some recent successes should be noticed (Makhnach et al., 2004; Currás et al., 2012) and promise new perspectives for tufa palynology (Capezzuoli et al., 2014). In their palynological study of the Bagnoli tufa in Tuscany (Fig. 1), Bertini et al. (2014) have recently demonstrated that pollen conservation are actually not limited by the alkalinity of water from which tufa precipitates. Investigation of depression deposits (lower energy) revealed well preserved and relatively abundant pollen with no differential preservation features whereas slope deposits (higher energy) have lower pollen concentration. The depositional energy of the environment during the deposition of tufa thus seems to play a more important role in controlling pollen concentration.

4. Geochronology

To unravel the timing of environmental changes estimated from those proxies, accurate chronologies of tufa archives are necessary. The various direct and non-direct methods available to date tufa are explored in the next section.

4.1. Direct dating on tufa (radiocarbon and U-series dating)

As tufas are mainly composed of calcite, they are suitable targets to be directly dated by radiocarbon and U-series methods. Those techniques cover complementary time periods: ^{14}C dating provides a chronology of Holocene tufa deposition whereas U-series can be use to date Pleistocene older sequences from MIS 5 (c. 125 ka) to MIS 11 (c. 420 ka).

For recent tufas, radiocarbon dating on carbonate appears to be the most suitable method. However, tufa deposition depends on physicochemical parameters as well as the activity of instream vegetation, algae and bacteria. Biogenic effects may introduce a significant discrepancy between the tufa calcite ^{14}C and atmospheric ^{14}C activity, as well as the so-called hard water effect (Broecker and Walton, 1959) which is due to dissolution of ^{14}C free carbonate from surrounding catchment that results in a lower ^{14}C activity of water bicarbonates than atmospheric CO_2 activity. Variation of this hard water effect along the tufa deposits is likely as a result of changes in environmental and/or climate conditions. Constraining the hard water effect is thus a prerequisite for a reliable ^{14}C derived chronology. Besides carbonate, tufa also encloses organic materials, charcoal, wood and cyanobacterial biofilm. Presence of the latter is not systematic and greatly depends on hydrodynamics: from very rare in highly energetic environment to frequent in quiet to limnic environments. Comparison of ^{14}C dating of tufa carbonate and enclosed organic matter can provide a good estimation of the apparent reservoir age, if the organic material is (1) proven contemporaneous of carbonate precipitation (no erosive input) and (2) clearly associated to atmospheric CO_2 (seed, stem or leaf for high-size superior plant; Hatté and Jull, 2013). Following this strategy, Pazdur et al. (1988a) estimate apparent reservoir age of Holocene tufa in different contexts, ranging from 910 to 3900 years. This very large range illustrates that transposition of an apparent reservoir age, estimated in a specific context, to another part of tufa has to be properly thought. Srdoc et al. (1980) also pointed out this fact and sent the clear message of a good knowledge of several factors peculiar to tufa formation, including the initial activity of fresh tufa.

For interglacials older than the Holocene, U-series dating is a strong tool to directly date tufa calcite. Recent analytical development such as multi collectors inductively coupled mass spectrometry (MC-ICP-MS) increase substantially the precision on U-series age determinations, allowing therefore the use of such advanced technique, to date very young $CaCO_3$ precipitations with high precision and sub annual resolution Shen et al. (2013).

Fig. 5. Conservation and abundance of malacofaunes in tufa. A and B: Mollusc shells preserved *in situ* at La Celle and C: screen overflow from the same tufa sequence (photos: N. Limondin-Lozouet).

The U-series dating method is based on the difference between the geochemistry of uranium and thorium in aqueous solutions (Ivanovich and Harmon, 1992). The high solubility of uranium in aqueous solutions allows uranium (U^{+6}) to be transported in oxygenated water, in contrast to the quasi-absent thorium (Th^{+4}), which has very low solubility in natural waters (Gascoyne, 1992).

While tufa calcite precipitates, a measurable trace of dissolved uranium (with the same $^{234}U/^{238}U$ activity ratio of the water) is incorporated but it is relatively devoid from insoluble thorium. After deposition, ^{230}Th is produced by the radioactive decay of ^{234}U, and the activity ratio $^{230}Th/^{234}U$ increases with time until radioactive secular equilibrium is re-established (~500 ka). If the radioactive system remains closed (*i.e.* there is no gain or loss of parent or daughter isotopes by any means except the radioactive decay), the activity ratios $^{230}Th/^{234}U$ and $^{234}U/^{238}U$ are thus only dependant on the time elapsed since the tufa precipitated. If the above conditions apply (pure calcite derived from physicochemical variations, perfectly closed system, etc.), the ages obtained by U-series disequilibrium correspond to calendar ages that do not require correction for atmospheric CO_2 variation and non-equilibration with atmospheric CO_2 during degassing, unlike ^{14}C ages.

However tufa is often porous and a detrital fraction may be trapped in its structure during or subsequent to deposition. This contamination is then indicated by the presence of non-radiogenic ^{232}Th in tufa deposits. Consequently, a part of ^{230}Th and uranium isotopes measured may be derived from the detrital materials. In this case, a correction should be made in order to estimate authigenic (original) uranium isotopes and ^{230}Th, used then to calculate the age of tufa (Ku and Liang, 1984).

In addition, closure of the radioactive system is a crucial but difficult to verify condition. Eikenberg et al. (2001) have demonstrated consistent and reliable ages for the spring tufa deposits of Hollgrotten (Switzerland) by combining two chronometers ($^{230}Th-^{234}U$ and $^{226}Ra-^{230}Th$). Using combinations of two or more dating chronometers may offer the possibility to detect any breakdown of the conditions of the closed system (i.e. $^{230}Th/^{234}U$ vs. $^{231}Pa/^{235}U$) and verify this assumption. In the case of discordant ages, processes that may have affected the sample such as U-uptake, leaching, etc. can be suggested (Cheng et al., 1998).

4.2. Other non-direct dating (palaeodosimetrc methods on associated material)

Palaeodosimetric methods (thermoluminescence-TL, optically stimulated luminescence-OSL, electron spin resonance-ESR) were also used to precise the ages of some famous Palaeolithic sites in Western Europe associated with interglacial tufa sequences. These different methods benefit from the low dose rates that characterise tufa sequences and, even if they are usually less accurate than radiometric methods, allow direct dating of human activities or geological events through the analyses of associated archaeological or mineralogical materials. For example, reliable ages were provided by thermoluminescence on burnt flints at Beeches Pit (Preece et al., 2007), or by ESR/U-series dates on palaeontological remains (tooth) at La Celle (Limondin-Lozouet et al., 2010). The ESR method has also been used to date quartz grains extracted from fluvial sediments underlying tufa sequences, for example at Longpré-les-Corps-Saints (Somme, France; Fig. 1; Laurent et al., 1998) or directly on the carbonate materials such as at Bilzingsleben and Ehringsdorf (Schwarcz et al., 1988).

Direct ^{14}C and U/Th dating on tufa and dating performed on the archaeological and palaeontological material or on interlying sediments must be combined with the stratigraphical and biochronological information, to allow building confident and detailed chronological framework to tufa deposition and the included archaeological levels.

5. Tufa geochemistry and climate reconstruction

5.1. Human, environment and climate (co-)evolution: decadal-scale geochemical analyses

The main advantage of tufa deposits is their potential for combining archaeological and environmental data with climate reconstructions from the geochemical analyses of calcite. Since the late 1980s, the most studied proxies in tufa are the oxygen and carbon stable isotopes in calcite (Pazdur et al., 1988b; Andrews et al., 1993, 1994, 1997).

At decadal-scale resolution (i.e. centimetric sampling resolution, continuously), tufa calcite oxygen stable isotopes ($\delta^{18}O$) have been shown to record the isotopic composition of regional rainfall, which in turn reflects mainly the mean annual air temperature in modern and Holocene tufas (Andrews et al., 1997). Parallel, carbon stable isotopes in tufa calcite ($\delta^{13}C$) were known to indicate moisture availability (linked to biomass type/abundance) and rainfall intensity (Andrews, 2006; Garnett et al., 2004). Recently, their suitability as climatic proxies for older Pleistocene periods has been demonstrated by comparing geochemical data to environmental reconstructions (mainly based on malacofaunes) from the same sequences at Caours and La Celle (Northern France; Dabkowski, 2011; Dabkowski et al., 2011).

Since the 2000s, there has been increasing interest in the so-called "trace element" proxies: Mg/Ca and Sr/Ca ratios in tufa calcite (Dabkowski et al., 2012; Garnett et al., 2004; Ihlenfeld et al., 2003). These ratios have been shown in tufa to mainly depend on rainfall availability through various processes in the aquifer. They are thus valuable complementary data to tufa $\delta^{13}C$ which is itself controlled by combined, hardly isolatable, parameters (Dabkowski et al., 2012; Garnett et al., 2004).

When archaeological remains are present in a tufa sequence, the isotope and trace element proxies thus allow determination of the decadal-scale evolution of both temperature and humidity around the time of occupation. Consequently, archaeological settlements can be positioned in a wider climatic framework by correlation with other regional or global climatic records (e.g. from lake deposits, ice cores, marine cores, etc.), even if such comparisons have to be very carefully discussed and must match with the whole set of environmental and chronological data available from a site (Garnett et al., 2004; Dabkowski, 2011). In this way, possible co-evolution of human behaviour with climate could be understood at a given site and general patterns of climate/society interactions be designed by combining data from several sites.

Investigations at Caours (MIS 5) and La Celle (MIS 11) indicate for instance that the main human occupation is contemporaneous at both sites with the interglacial climatic optimum, i.e. temperature and humidity maxima recorded by geochemical proxies, which is highly coherent with the parallel malacological study that shows the simultaneous maximal development of forest environments (Dabkowski et al., 2011, 2012). Moreover, Caours Middle Palaeolithic and La Celle Lower Palaeolithic levels are both preserved in fine greyish tufa deposits (palaeosoils) in a perched springline tufa system (Antoine et al., 2006; Limondin-Lozouet et al., 2006, 2010). Those preliminary observations thus show similar human–environment–climate interactions during the Eemien and the MIS 11. This example demonstrates that tufa geochemical investigations must be integrated to multidisciplinary studies to provide relevant information on human occupation contexts.

5.2. Contribution of tufa studies to climate investigation at a "human" time scale: high-resolution geochemical analyses

In many recent to Pleistocene tufa formations, very well crystallised, strongly laminated deposits are preserved. They result from calcite accretion which encircles a mobile nucleus rolling on the channel or pool floors (oncolithes), expands around vegetation branches, trunks or stems (tubular stromatolites), or simply covers level surfaces run through by intermittent/weak flows (Fig. 6). Laminae have been demonstrated to mostly reflect seasonal climatic and environmental variations (Chafetz and Folk, 1984; Freytet and Plet, 1991; Freytet and Verrecchia, 1998). Recent studies of sub-annual isotopic variability in actively depositing (modern) tufas have shown the large potential of such laminated deposits as archives of high-resolution climatic data (Hori et al., 2008; Kawai et al., 2009) and encouraged investigation of fossil deposits. At infra-lamina sampling resolution, calcite $\delta^{18}O$ in Holocene to Pleistocene laminated tufas thus appears to track seasonal variations of water temperature (Matsuoka et al., 2001; Andrews and Brasier, 2005; Brasier et al., 2010; Dabkowski et al., submitted for publication).

Tufas thus provide annual to infra-annual climatic data significant at a "human" time scale, comparable to high-resolution records from tree rings or mammal teeth. The challenge is to establish correlation between tufa seasonal records, mammal data, and the phases of human occupation, especially when these are short (seasonal) or repetitive. Contemporaneity could be achieved at high resolution when archaeological artefacts are actually encrusted in laminated tufa or when precise stratigraphical markers are present (volcanic ashes, fire events, etc.). In the other cases, such correlations would be limited by dating precision and appear nearly impossible to manage with sufficient confidence in older deposits.

6. Possible contributions of tufa investigation to European archaeology

In Northwestern Europe, fluvial areas have been favoured for human settlements since the Early Palaeolithic (Antoine et al., 2003, 2010; Ashton et al., 2006; Bridgland et al., 2006). For these oldest periods, well preserved archaeological sites with good chronological data are sparse. Recently, old Acheulean lithic assemblages including handaxes dated between 700 and 600 ka have been identified in fluvial terrace context in the Loire Basin (Centre of France) and Southern England (Parfitt et al., 2005; Despriée et al., 2007, Moncel et al., 2013). During the Middle Pleistocene, Acheulean settlements are specifically recorded during temperate periods, especially MIS 11 and 9 interglacials, or during transitional contexts from interglacial to glacial (Antoine et al., 2003; Nicoud, 2011). From c. 300 ka, Middle Palaeolithic sites are also preserved in interglacial (MIS 7 and 5) or transitional (MIS 7/6) contexts whereas little or no settlements are associated to MIS 6 and 2 full glacial conditions (Antoine et al., 2003). Thanks to their specific distribution both in space (associated to fluvial context) and time (specific to interglacial context), tufa are very likely to document key periods of the human biological and societal evolution in Europe through multidisciplinary investigations.

6.1. Significance of Pleistocene tufas to human evolution and society studies

The MIS 11 (*c.* 400 ka) and 5e (Eemian; *c.* 125 ka) interglacials are especially well represented in European fluvial systems as they experienced very wet and warm climatic conditions favouring tufa precipitation (Pentecost, 1995, 2005). Together with the Holocene, these past interglacials are also key-periods for human biological and societal evolution in Europe.

Data from the fields of human palaeontology and palaeogenetics together estimate the separation of African and European lineages leading to *Homo sapiens* and *H. neanderthalensis* at *c.* 400 ka (Hublin, 2009). As one of the longest (>30 ka) and warmest past interglacials, the MIS 11 experimented climatic conditions which could have favoured the spread of human populations over European middle latitudes. Isolation of these areas during the next cold periods then partially explains the individualisation of European Neandertalian populations (Hublin, 2009). Tufa potential to preserve human fossils, such as the skull of *H. heidelbergensis* discovered in Bilzingsleben (Germany; Mania et al., 1994), might be explored to document this stage of human evolution and its environmental and climatic context.

From MIS 11 too, controlled fire use appears widespread over Northern Europe and allows significant modifications in natural resource and territory management, parallel to an increase in human social/intellectual capability (Gowlett, 2006; Roebroeks and Vila, 2011; Rolland, 2004). A prime example indicating fire control and social activity around fires is Beeches Pit (United Kingdom), where a structured series of hearths associated with fractured bones and lithic artefacts has been identified in the upper part of a tufa sequence (Fig. 1; Preece et al., 2007). Fine tufa texture favoured *in situ* preservation of burnt sediments and artefacts as for many other ancient fire use vestiges (e.g. Bilzingsleben II – MIS 11- and Weimar Ehrindorf – MIS 11, Germany; Gowlett, 2006; Mania, 1991; Vértesszöllös travertine, Hungary; Pentecost, 2005; Roebroeks and Vila, 2011).

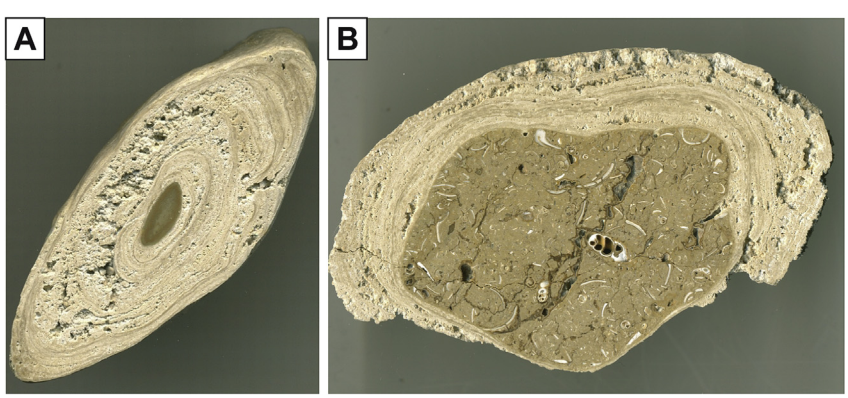

Fig. 6. Laminated tufa: oncoliths from Changis-sur-Marne (Seine Valley; see Fig. 1) formed around two different kind of nucleus. A. flint nucleus; B. limestone nucleus (photos: P. Antoine).

Emergence of the modern cognition and associated innovations is currently debated, with crucial questions on the time and rhythms of this process and so, whether or not it is unique to *H. sapiens*. Appearance of "cultural modernity" in Europe is commonly associated with the arrival from Africa of anatomically modern humans around 40 ka. However, some authors suggest that material cultures similar to those of the Upper Palaeolithic could have gradually appeared in Europe around 100–120 ka, favoured by demographic and societal changes induced themselves by climatic variations and especially the MIS 5 optimum more pleasing conditions (d'Errico and Stringer, 2011; Pellegrin and Soressi, 2007). Last Interglacial tufa formations appear as a great opportunity to precisely document climate and environmental variations occurring during this transitional cultural time and will thus contribute to the debated question of the appearance of "cultural modernity".

Besides, the occupation of northwest Europe during the Eemian Interglacial is yet poorly documented. However, the Caours sequence in Northern France (Somme Basin) includes at least for major Middle Palaeolithic levels contemporaneous to the MIS 5e (Antoine et al., 2006; Locht et al., 2009). Thanks to the excellent preservation of bone and lithic artefacts in tufa context, valuable information regarding to human processing features on large mammals, hunt pattern or lithic industry were highlighted in Caours. Additionally, as tufa allows *in situ* conservation, spatial investigation to define activity areas and investigate interaction between them have been initiated (Locht et al., 2009). The multidisciplinary study of the site has also allowed a well documented reconstruction of the climatic and environmental condition prevailing at the time of the human occupations (Antoine et al., 2006; Dabkowski, 2011; Dabkowski et al., 2011; Locht et al., 2009). The study of Caours therefore underlines the importance of fluvial sediments, and in particular tufa deposits for the preservation of interglacial Palaeolithic sites.

6.2. Holocene tufas as records of anthropisation/impact of human activity

Our modern interglacial, the Holocene, obviously allowed emergence of new cultural and economical behaviour through the so-called "neolithisation". Fluvial systems including tufas appear highly responsive to the environmental variations induced by human activities and especially land cleaning and planting, originally by streams in the valleys but rapidly extending (Antoine et al., 2002; Limondin-Lozouet et al., 2002; Pastre et al., 2001, 2002).

From 5 to 6 ka, calcareous tufa formations appear progressively scarcer and less extended whereas they were previously widely represented and well developed during the Boreal and the early Atlantic stages (Pentecost, 2005). The factors leading to the European "late-Holocene tufa decline" (Goudies et al., 1993) are still widely discussed and compilation of precise stratigraphical, chronological and archaeological data are needed to properly identify and understand what happened. Anyway, most of the authors agree that this decline is the result of combined effects of the climatic change at the end of the Atlantic and the increasing anthropogenic impact on fluvial environments throughout Protohistory (Geurts, 1976; Pentecost, 1995, 2005; Vaudour, 1986).

Calcareous tufas are indeed highly responsive to environmental modifications, just as any fluvial systems (Antoine et al., 2002; Pastre et al., 2001, 2002). Increasing forest clearance to obtain agricultural lands both (1) reduces surface of well developed forest soils and the resulting acidity of groundwaters which would lead to decreasing calcite dissolution in the aquifer and so, less calcite (re) precipitation as tufa, and (2) allows the increase of erosion processes, and consequently of the detrital charge of rivers, which would damage or reduce tufa deposits. Where tufa deposition continued in the early Holocene, continuation of closed forest conditions is recorded. For example, the mollusc assemblages preserved in the Direndall tufa (Kopstal, Mamer Valley, Luxemburg) are dominated by shade-loving taxa and show prevalence of natural forest environments which allow tufa deposition until at least 900 years BP (Meyrick, 2002). However, an episode of expansion of taxa favouring fairly open environments is observed in the upper part of the tufa (c. 1640 $\pm$ 60 yrs BP) and seems associated with a general decrease of the tufa deposition rate. Extensive archaeological evidences indicate Roman presence in the surrounding area, involving deforestation to obtain agricultural land from this time (Ternes, 1991; Polfer and Thiel, 1996). These observations strengthen the hypothesis that deforestation must be a major factor leading to the tufa decline during the Holocene (Meyrick, 2002; Preece, 1980). Distribution and development of calcareous tufas through the Holocene thus appears as a promising new indicator of the anthropisation and impact of human activity on natural environments.

A particular kind of tufa deposits developing parallel to man-built water-related edifices such as aqueducts, irrigation structures, baths, and more recently, industrial boilers, water pipes, etc. must be also mentioned here. Investigations of "man-made" tufas indeed contribute in various ways to Antiquity to industrial archaeology. Roman aqueducts present the largest potential as more than 1300 of them are preserved, covering thousands of km over the ancient Roman Empire, in Europe and around the Mediterranean (ROMAQ Project Database, 2012). The arrangement and geometry of deposits, including decadal, annual or seasonal laminations, phases of non-deposition or cleaning out events, may inform on the duration and dynamic history of an aqueduct, especially if combined with appropriate dating. Several studies of these kinds have been conducted on Southeastern France aqueducts (i.e. Nîmes, Fréjus, Arles) and provided valuable information for the understanding of the construction, maintenance and abandonment of these arrangements (Guendon et al., 2002; Guendon and Leveau, 2005; Leveau, 2004). Geochemical analyses can also be performed on the laminated tufa calcite, providing supplementary information on aqueduct history, water quality (which could track pollution) and sources, and climatic parameter evolution (Gilly, 1986; Sürmelihindi et al., 2013).

7. Synthesis and conclusion

This short overview of human–environment–climate interactions during interglacials from the Lower Palaeolithic in Europe highlights the importance of fluvial areas and especially calcareous tufas in European archaeology. Calcareous tufas are specific deposits with many characteristics allowing development of investigations in various fields from archaeology, palaeontology (including palaeoanthropology) to past environment and climate study, in well dated contexts.

1) Tufas are very likely to preserve *in situ* archaeological levels with an excellent conservation of lithic artefacts, hunted large mammal remains or even fire hearts or burned areas. Fast accumulation of calcite could rapidly sealed human occupations after their abandonment which allows investigating artefacts spatial organisation to identify activity areas and their possible interactions within a site. Perched sprinline tufa systems on the slopes and river banks are more suitable for human settlement then preservation of archaeological levels. In those systems, palaeosoils are common, characterised by fine greyish tufa deposits showing low deposition features. Where archaeological levels are uncovered in tufa, they are always preserved in such contexts. These greyish tufa palaeosoils are generally

contemporaneous with maximal development of forest environments and the resulting stabilisation of the surrounding area, recorded by bioindicators (especially malacofaunes), and with the climatic optimum observed in geochemical proxies.

2) The wide variety of palaeontological remains preserved in tufas provides a strong support to environmental investigations. Molluscs are particularly well represented all along tufa sequences both in quantity and quality. They are probably the most suitable bioindicator to investigating long term environmental variations in tufa. Additionally, they can also be reliable chronological indicators.

   Large mammal remains are very common in tufa deposits and usually preserved in anatomical connection or as left by humans. Together with other less frequent vertebrate fossils (small mammals, amphibian, reptiles, birds, fishes), they support palaeontological and environmental investigations, as well as archaeozoological studies when human processing features (i.e. cut marks or breaking patterns) are observed. In some exceptional case, human remains, generally cranial bones, were also uncovered in tufa providing valuable material to the investigations on human evolution in Europe.

   Plants are also widely represented in calcareous tufa, mainly as imprints of rapidly encrusted leaves or fruits that have then decayed. High quality preservation of external details generally allows specific determination and thus supports environment investigations. Pollen is generally rare in tufa sequences but recent investigations show it might be preserved in low energy (depression) deposits. Palynological studies of tufa should be thus developed to be integrated into multidisciplinary environmental investigations combining all the bioindicators available in tufas.

3) As they are mainly comprised of calcite, calcareous tufas are also suitable for geochemical studies allowing climate reconstructions at various time scale (from decadal to seasonal resolution). Stable isotopes ($\delta^{18}O$ and $\delta^{13}C$) and trace element ratios (Mg/Ca and Sr/Ca) have been recently demonstrated to be reliable proxies of temperature and humidity (mainly rainfall intensity) variations in Pleistocene tufas just as in Holocene formations.

4) Tufa calcite is a suitable target for direct dating by the radiocarbon and U-series methods. Those techniques allow covering complementary time periods: ^{14}C dating can be used to date Holocene tufa deposits whereas U-series is suitable for Pleistocene interglacial sequences. Additionally, various materials comprised in tufa (e.g. tooth, bone, burnt flint) and associated fluvial sediments (from which quatz grain can be extracted) can be dated by palaeodosimetric methods such as thermoluminescence (TL), optically stimulated luminescence (OSL) or electron spin resonance (ESR). These various methods must also be combined with stratigraphical and biochronological information. Confident and detailed chronological framework can thus be built for tufa deposits and the included archaeological levels.

Calcareous tufas are therefore a unique continental archive that can provide very detailed and interconnected archaeological, environmental, climatic and chronological data. We believe that integrative multidisciplinary approaches on tufas must be encouraged, combining all data available at a site with systematic search for archaeological artefacts, paying attention to their spatial organisation, and for all associated environmental indicators. Tufas actually emerge as a new exciting field of investigation to address some major archaeological issues in Europe into a well documented environmental, climatic and chronological framework.

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